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INVESTIGATION OF THE DENSITY OF GRANULAR FOAM-GLASS CERAMIC BY MATHEMATICAL MODELING

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The possibility of producing granular foam glass by using household and commercial cullet, low-melting ceramic filler and organic additives is examined. An ecologically safe resource-conserving technology has been developed for obtaining heat-insulating material — granular foam-glass ceramic. Mathematical modeling of an experiment is used to investigate the physical and technical characteristics of the granules.

Key words: cullet, foam-glass ceramic, granules, density, temperature, organic matter, variational factor, optimization criterion, planning matrix, dispersion, mathematical model.

Russian industry can be supplied with highly efficient heat-insulation materials most quickly and effectively by developing and adopting modern science-intensive technologies for producing foam glass from aluminum silicate compounds.

The main advantage of foam glass over the well-known heat-insulation materials lies in the unique combination of heat-insulation and construction properties, which makes it possible to use it in different areas of industry. However, foam glass is not widely used because the production process is quite complex and energy-intensive and the degree of automation of the technological process and control is low [1].

The improvement of the foam-glass production technology using optimized compositions and technological processes with adoption of an automatic production control system is one of the important problems whose solution will make it possible to improve quality, significantly reduce energy consumption and increase the utilization range and volumes of the material [2].

Granular foam-glass ceramic, developed by Tomtekhnologiya JSC (patent No. 2374191), was the object of study. The content of the rational composition was cullet 67 – 84%, low-melting ceramic filler 8 – 25%, organic additives 3% and foaming agent 5%.

Heat-treatment was conducted at temperature 830°C and the foaming time was 5 – 15 min. The annealing of the granules was combined with the process of cooling the furnace. Annealing is conducted in order to remove the residual thermal stresses which can give rise to cracks and fracturing of the granules.

The following parameters of the physical-technical characteristics of the granules were obtained as a result of the experiments:

– mechanical strength in compression 0.82 – 2.5 MPa;
– thermal conductivity 0.067 – 0.087 W/(m · K);
– average density 200 – 290 kg/m^3;
– water absorption 3.2 – 2.6%.

The experiments on the development of a technology for the production of granular foam-glass ceramic are laborious and expensive. In this case, in order to determine the optimal physical and technical parameters, compositions and technological processes it is expedient to use the full factorial experiment method (FFE). This substantially reduced the time and material resources required to perform the investigations.

The methods of modeling with the aid of a mathematical model expressed by an equation representing the process were implemented in the Mathcad system. The variational factors, variational interval and the upper and lower levels of the factors were chosen on the basis of the problem posed and a priori data.

The density (in kg/m^3) of the granules of foam-glass ceramic at 20°C was chosen as optimization criterion Y.

The objective of the experiment was to investigate the effect of the following factors on the average density of the granules:

X_1) content by weight of the low-melting ceramic filler, %;

X_2) foaming time (at t = 830°C), min;

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TABLE 1. Basic Level and Variational Intervals of Factors

Factor	X_1	X_2	X_3
Basic level	10	10	6
Variational interval	5	5	3
Upper level	15	15	9
Lower level	5	5	3

X_3) content by weight of the separation medium, which keeps the granules from sticking to one another during heat-treatment, in the granules during annealing in a rotary furnace, %.

The experiments were performed using batch with the following composition: glass powder, low-melting ceramic filler, organic additive and coke. The content of coke and organic additives is constant in all experiments.

The components of the batch were metered out and loaded into a 75T-DrM rod vibratory mill, where mechanical activation was performed to specific surface area 4000 cm^2/g. A PSKh-2 apparatus was used to determine the dispersity of the batch (according to the impermeability of the layer of powder). The batch obtained was mixed with water. After the required plasticity was reached, granules 10 mm in size were formed from the ceramic body.

The variational interval and step for each of the independent variables is determined by the problem of taking all possible variants into account to the maximum extent possible (Table 1).

Three factors (X_1, X_2, X_3) were chosen to plan the experiments. The model is an incomplete quadratic model.

A sequence for performing experiments was obtained as a result of randomization using a table of random numbers. The total number of experiments was 24.

Analysis of the experimental results gave a linear mathematical model of the form

$$Y = 285.9 - 20.0X_1 - 17.5X_2 + 22.1X_3 + 1.02X_1X_2 + 8.2X_1X_3 - 1.62X_2X_3. \quad (1)$$

Cochren's criterion, fond to be 0.41, was used to check the uniformity of the variances.

The hypothesis of significance of the regression coefficient was checked by means of Student's criterion t, which was found to equal $t = 2.12$. The variance of the adequacy of the model was 1.13.

The hypothesis of model adequacy was checked with the aid of Fisher's F criterion. The computed value was $F_{comp} = 0.42$; the tabulated value is $F_{table} = 4.49$. Since $F_{comp} < F_{table}$, the hypothesis of model adequacy is accepted.

Interpreting the results, it can be concluded that all linear effects are significant, i.e., all factors investigated (the content of low-melting ceramic filler, foaming time and content of the separation medium) have a substantial effect on the average density of the granules of foam-glass ceramic. It is evident from the equations that X_3, the content by weight of the separation medium, has the greatest effect on the change of the response function Y (the coefficient of X_3 is $b_3 = 22.1$); X_1, the content by weight of the low-melting ceramic filler in the batch, is the next most influential ($b_1 = -20.0$), followed by the foaming time X_2 (at $t = 830°C$), min ($b_2 = -17.5$).

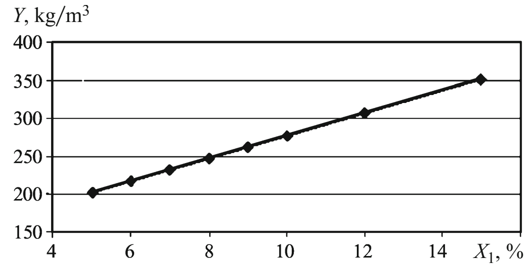

Fig. 1. Criterion Y versus X_1 for X_2 = const and X_3 = const.

In summary, the series of coefficients of regression for linear effects can be ordered by modulus as follows: $b_3 > b_1 > b_2$. The sign of the coefficients b indicates how a factor influences an experimental result.

The dependences Y versus X_1 for X_2 = const and X_3 = const are shown in Fig. 1.

In Fig. 1, obtained from the equation of the mathematical model, where the factors X_2 and X_3 were taken to be constants, the dynamics of the increase in the response function Y with X_1 increasing from 5 to 15% is shown clearly. The low-melting ceramic filler X_1 is an inhibitory factor of the pore-formation process and the foaming time. The formation time of the liquid phase is increased and lags behind the period of intense gas-formation. As a result, a large part of the gases does not participate in the foaming process and freely exceeds the limits of the samples. For clay content by weight from 5 to 15% the density of the granules fluctuates from 200 to 340 kg/m^3. The optimal average density 200 – 260 kg/m^3 is reached with content 8 – 10% of the low-melting ceramic filler in the batch. The investigations showed that these samples possess relatively high mechanical strength 1.7 – 2.2 MPa. For comparison, in the case of keramzit with mechanical strength in compression 0.6 – 5.5 MPa the density is 500 – 600 kg/m^3.

The mathematical model was used to construct a plot (Fig. 2) of the average density Y versus the foaming time X_2 (in min) for $t = 830°C$ and constant factors X_1 and X_3.

The average density of the granules decreases with increasing foaming time for constant X_1 and X_3. For foaming time 5 min the density of the samples is 289 kg/m^3 and reaches 237 kg/m^3 at foaming time 8 min. By increasing the foaming time it is possible to regulate and bring into correspondence the formation time of the glassy phase with the period of the most active degassing process. The samples obtained with the longest foaming time possess a fine and brittle interpore barrier. The number of pores decreases but their size increases many-fold. The pores decrease in number but increase many-fold in size. Dissipative phenomena — pore

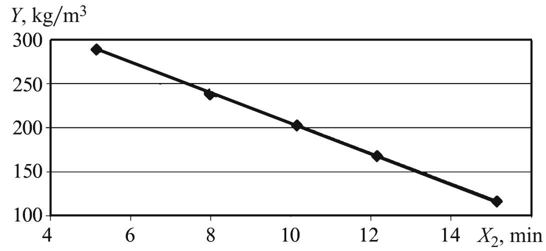

Fig. 2. Criterion Y versus X_2 for X_1 = const and X_3 = const.

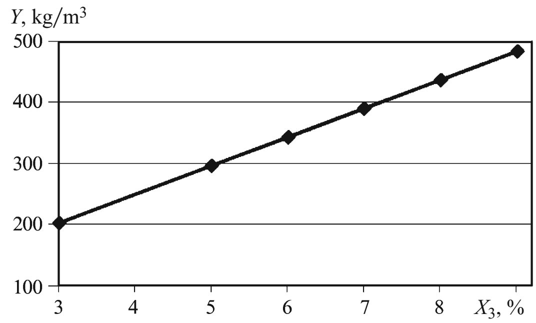

Fig. 3. Criterion Y versus X_3 for X_2 = const and X_1 = const.

growth and collapse — are more likely to occur. The investigations of the mechanical strength of the samples in compression showed that for the samples obtained with foaming time 5 min the mechanical strength is 1.7 MPa. As the foaming time increases to 12 min, the mechanical strength in compression decreases to 0.8 MPa. It follows from an analysis of the equation that the optimal variant is foaming time from 5 to 8 min, for which the average density lies in the range 237 – 289 kg/m^3.

The dependence of the average density Y on the content by weight of the separation medium X_3 (%) for constant factors X_1 and X_2 is shown graphically in Fig. 3. For the amount of separation medium 3% (lower limit) the density of the granules is 202.6 kg/m^3 and reaches 484.0 kg/m^3 at the upper limit 9%. For average density in the range 200 – 300 kg/m^3 the content of the separation medium is 3 – 5%.

Analyzing Eq. (1) and the plots in Figs. 1 – 3 and setting the density 200 – 300 kg/m^3, we determine the optimal parameters characterizing the formation of granules of foam-glass ceramic.

Optimal Formation Parameters for Foam-Glass Ceramic Granules

Density, kg/m^3	200 – 300
Content of low-melting ceramic filler, wt.%	8.0 – 10.0
Content of separation medium, wt.%.	3 – 5
Sintering interval, min (foaming time)	5 – 10
Foaming temperature, °C	830

A quadratic mathematical model of the technological process of the production of the granules of foam-glass ceramic was constructed with the aid of a multifactorial experiment to determine the quantitative differences between the results of the investigations. The values and interval of variation of the factors X_1 and X_3 were reduced.

TABLE 2. Basic Level and Variational Intervals of the Factors

Factor	X_1	X_2	X_3
Basic level	4	10	4
Variational interval	2	5	1
Upper level	6	15	5
Lower level	2	5	3

On the basis of a series of screening experiments we determine the factors that have the greatest influence on the response function Y — the average density of the granules (kg/m^3).

Three such factors were found:

X_1) foaming time (at t = 830°C), min;

X_2) content of low-melting ceramic filler, wt.%;

X_3) content of the separation phase in the granules after annealing, wt.%.

The basic level, variational interval and the upper and lower limits of the factors are indicated in Table 2.

After analyzing the experimental results using the planning matrix developed, we obtained a quadratic mathematical model expressed by the following equation:

$$Y = 255.04 - 6.1X_1 + 9.42X_2 + 15.1X_3 - 8.25X_1X_3 - 8.5X_2X_3 - 10.5X_1X_2X_3 + 21.18X_2^2 + 13.4X_3^2. \quad (2)$$

The computed Fisher criterion for this equation equals $F'_{\text{comp}} = 1.795$; the tabulated value is $F'_{\text{table}} = 2.06$.

Since $F'_{\text{table}} > F'_{\text{comp}}$, the equation adequately describes the process. Using the mathematical model, expressed by the equation, that adequately describes the technological process, we construct a 3M plot for different grouping variable factors.

The methods for modeling with the aid of a mathematical model, expressed by the equation of the process, were implemented in the Mathcad system. The wide choice of the 3M plots constructed made it possible to obtain the optimal plots (for displaying the results) of the surface with a description by a quadratic functional of the dependence of the change in the properties of the ready material.

The optimal variants characterizing the change in the properties of the granules are presented in the 3M plots (Figs. 4 and 5). The spatial plots represent the surfaces described by a quadratic function with variable properties of the ready material. The 3M representation supports an interactive rotation, change in proportions and perspectives. The limiting values of the average density 228 – 1056 kg/m^3 were obtained by varying the factors X_1 = 2 – 6 min, X_2 = 5 – 15% and X_3 = 3 – 5%.

Figure 5 displays 3M plots where the response function Y, the average density (in kg/m^3), depends on the changes in

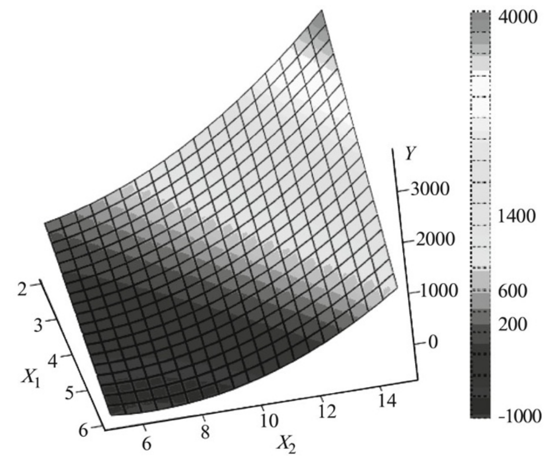

Fig. 4. The average density Y of foam-glass granules versus the foaming time X_1 and content of the low-melting ceramic filler X_2 in batch with separating medium content 4%.

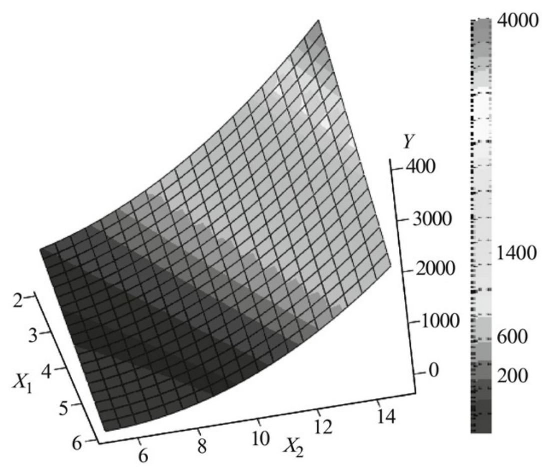

Fig. 5. Average density Y of the foam-glass ceramic granules versus the foaming time X_1 and content (by weight) of the low-melting ceramic filler in batch with separating medium content 3%.

X_1, the foaming time, the content (by weight) of the low-melting ceramic filler X_2 in the batch and the content (by weight) of the separation medium X_3 = const. Analyzing the plots (Fig. 5) we can say that for variation of the content of the low-melting ceramic filler from 5 to 15%, the foaming time from 2 to 6 min and content of the separation phase equal to 4% the average density was 228 kg/m^3.

For elevated content of the low-melting ceramic filler X_2 in the batch up to 10%, reduced content of the separation medium to 3% and foaming time remaining unchanged at 4 min the average density of the process increased and was equal to 1056.74 kg/m^3 (Fig. 5).

The effects of the interaction constitute the nonlinear part of Eq. (2). The meaning of this effect is that the action of a

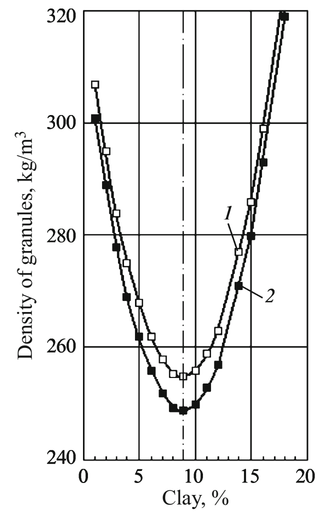

Fig. 6. Density of foam-glass ceramic granules versus the clay content in the batch.

factor depends on the level occupied by the other factor. By changing the independent variables in proportion to the values of the regression coefficients we move in the direction of the gradient of the response function along the shortest path to the optimum. As a result we obtain the plots presented in Fig. 6. The optimal content of clay is 9.8%, giving an average density of the granules 255.84 kg/m^3.

Analyzing the equations and plots obtained we can conclude that the introduction of 84 – 82% cullet, low-melting ceramic filler from 8.0 to 10.0%, organic additives 3% and coke 5% and taking the optimal foaming time for the granules 4 min results in a reduction of the average density to 228 – 265.58 kg/m^3.

In summary, the use of a full factorial experiment and constructing a quadratic mathematical model with mathematical analysis of the experimental results made it possible to determine a rational technological regime for fabricating granular foam-glass ceramic.

REFERENCES

1. A. A. Ketov, G. B. Ketova, A. I. Puzanov, et al., "Cullet as raw material for obtaining heat-insulation materials," *Ékologiya Prom-st' Rossii*, No. 8 (2002).
2. V. A. Lotov, "Promising heat-insulation materials with rigid structure," *Stroit. Mater.*, No. 11, 8 – 9 (2004).
3. I. G. Zadgenidze, *Planning of Experiments for Investigation of Multicomponent Systems* [in Russian], Nauka, Moscow (1976).
4. A. S. Apkar'yan, V. G. Khristyukov, and G. V. Smirnov, "Heat insulation of high-temperature heating units," *Steklo Keram.*, No. 1, 18 – 21 (2010); A. S. Apkar'yan, V. G. Khristyukov, and G. V. Smirnov, "Heat insulation of high-temperature heating units for ceramic production," *Glass Ceram.*, **67**(1 – 2), 52 – 55 (2010).